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On completeness of Born-Huang invariance conditions

S. K. SARKAR AND S. SENGUPTA

Solid State Physics Research Centre, Presidency College, Calcutta

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1. INTRODUCTION

Some authors (Lax 1965, Gazis & Wallis 1966, Keating 1966) have shown that the Born-Huang (B-H) translational rotational invariance conditions (Born & Huang 1954) are not sufficient and obtained additional rotational invariance conditions

The B-H conditions are derived for a finite lattice by considering the invariance of potential energy under translation and under infinitesimal rotation upto terms linear in rotation parameter (Liebfried & Ludwig 1960 1961 Hedin 1960) Lax (1965) considers the invariance of potential energy of a finite monatomic linear chain with nearest neighbour interaction (n.n) upto terms second order in rotation parameter and obtains additional conditions. Thus he shows that there is one independent n.n force constant for the linear chain while Born's theory gives two n.n. force constants

The B-H conditions are obtained for an infinite lattice by considering the transformation properties of coupling parameters (c.p.'s) under translation and rotation (Born & Huang 1954) However Gazis and Wallis (1966) and Keating (1966) obtain for an infinite lattice additional conditions following the same procedure as Lax. Hence they show that for a simple cubic primitive lattice there is only one independent n.n. force constant while Born's theory allows two such ones

In all these works harmonic potential energy is considered with first order c.p.'s taken as zero at equilibrium. Hence additional conditions involve second order c.p.'s (force constants) only The full equilibrium conditions of vanishing of stresses are not considered. Also they cannot assert whether their results are sufficient for rotational invariance and that other conditions do not exist.

Our aim in the present work is to examine critically this problem We find that it is necessary to discuss the purely translational and rotational invariance conditions separately from equilibrium conditions. The former conditions are valid even if the crystal is not in equilibrium. Only then we can check if a set of invariance conditions is complete or not. In the above mentioned works rotational invariance and equilibrium conditions are mixed up If however the equilibrium conditions are not mixed up we show that the usual set of B-H conditions are complete.

2. Translational and Rotational Invariance Conditions

Let us consider a finite lattice consisting of N particles and see whether the usual set of B-H invariance conditions are complete or not. If the completeness holds for the finite lattice for any value of N then it follows that the completeness is valid for an infinite lattice as well.

When there is no external field the lattice potential energy is a function of $3N-6(N>2)$ independent internal coordinates, such as distances and angles between the particles, which are invariant under translation and rigid rotation. Let these co-ordinates be denoted by $q_1, q_2 \ldots$ etc. If we expand the potential energy Φ about any configuration defined by the values $Q_1, Q_2, \ldots$ etc. of these co-ordinates we have

$$\Phi = \Phi_0 + \sum_k \frac{\partial \Phi}{\partial q_k} dq_k + \frac{1}{2} \sum_{kl} \frac{\partial^2 \Phi}{\partial q_k \partial q_l} dq_k \, dq_l + \ldots \qquad \ldots (1)$$

where $k, l = 1, 2, \ldots, (3N-6)$, and the coefficients are taken at the initial configuration $(\ldots Q_k \ldots)$. Since the co-ordinates are all independent, in general, all the coefficients of different orders are independent. There are, therefore, $(3N-6)$ independent first order coefficients and $(3N-6)(3N-5)/2$ independent second order coefficients.

The $3N-6$ q_k's can be expressed in terms of the $3N$ position co-ordinates $\boldsymbol{r}_1, \boldsymbol{r}_2 \ldots$ of the N particles. So Φ can be considered as a function of the $3N$ variables $\boldsymbol{r}_i$. If $\boldsymbol{C}$ be the position vector of the centre of mass and θ denotes collectively the three Eulerian angles then $(\ldots q_k \ldots, \boldsymbol{c}, \theta)$ and $(\ldots \boldsymbol{r}_i \ldots)$ define two alternative sets of $3N$ co-ordinates. Let the initial configuration $(\ldots Q_k \ldots \boldsymbol{C}, \theta)$ corresponds to $(\ldots \boldsymbol{R}_i \ldots)$. Then an alternative expansion of Φ about $(\ldots R_i \ldots)$ is the usual Born expansion

$$\Phi = \Phi_0 + \sum_{i\alpha} \Phi_1 \begin{pmatrix} i \\ \alpha \end{pmatrix} u_{i\alpha} + \frac{1}{2} \sum_{ij, \alpha\beta} \Phi_2 \begin{pmatrix} ij \\ \alpha\beta \end{pmatrix} u_{i\alpha} \, u_{j\beta} + \ldots$$

where

$$\boldsymbol{u}_i = \boldsymbol{r}_i - \boldsymbol{R}_i; \qquad \ldots (2)$$

$$\Phi_1 \begin{pmatrix} i \\ \alpha \end{pmatrix} = \left(\frac{\partial \Phi}{\partial r_{i\alpha}} \right) (\ldots \boldsymbol{r}_i \ldots) = (\ldots \boldsymbol{R}_i \ldots)$$

$$\Phi_2 \begin{pmatrix} ij \\ \alpha\beta \end{pmatrix} = \left(\frac{\partial^2 \Phi}{\partial r_{i\alpha} \partial r_{j\beta}} \right) (\ldots \boldsymbol{r}_i \ldots) = (\ldots \boldsymbol{R}_i \ldots)$$

$i, j = 1, 2, \ldots, N$, and $\alpha, \beta = 1, 2, 3$ are the cartesian component indices. There are $3N$ first order c.p.'s $\Phi_1\binom{i}{\alpha}$ and $3N(3N+1)/2$ second order c.p.'s $\Phi_2 \begin{pmatrix} ij \\ \alpha\beta \end{pmatrix}$.

Since the q_k's are internal co-ordinates the invariance under translation and rotation of any term in expression (1) is obvious But in expression (2) this is not so. An arbitrary choice of the c.p.'s will violate the invariance of Φ. So to guarantee the invariance the c.p.'s in (2) must satisfy certain conditions between themselves.

If we use the transformation

$$dq_k = \sum_{i,\alpha} \frac{\partial q_k}{\partial r_{i\alpha}} u_{i\alpha} + \tfrac{1}{2} \sum_{ij,\alpha\beta} \frac{\partial^2 q_k}{\partial r_{i\alpha}\partial r_{j\beta}} u_{i\alpha}u_{j\beta} + \cdots \qquad \ldots (3)$$

then expression (1) can be written in an equivalent form as (2) Comparing the two expression we obtain

$$\Phi_1 \begin{pmatrix} i \\ \alpha \end{pmatrix} = \sum_k \frac{\partial \Phi}{\partial q_k} \frac{\partial q_k}{\partial r_{i\alpha}}$$

$$\Phi_2 \begin{pmatrix} ij \\ \alpha\beta \end{pmatrix} = \sum_{kl} \frac{\partial^2 \Phi}{\partial q_k \partial q_l} \frac{\partial q_k}{\partial r_{i\alpha}} \frac{\partial q_l}{\partial r_{j\beta}} + \sum_k \frac{\partial \Phi}{\partial q_k} \frac{\partial^2 q_k}{\partial r_{i\alpha}\partial r_{j\beta}} \qquad \ldots (4)$$

We see therefore that the $3N$ first order c.p.'s in Born's expansion can be expressed in terms of $3N-6$ first order coefficients of expression (1) and the $3N(3N+1)/2$ second order c.p.'s in Born's expansion can be expressed in terms of $(3N-6)$ first order and $(3N-6)(3N-5)/2$ second order coefficients of eq. (1) Thus it follows that the $3N$ first order c p.'s must satisfy six relations among themselves and $3N$ and $3N(3N+1)/2$ first and second order c.p.'s together will satisfy another $3N(3N+1)/2 - (3N-6)(3N-5)/2 = 18N-15$ relations among themselves These are the conditions which guarantee invariance of (2) upto terms second order in u_i's.

Let us write the usual B-H conditions for a finite lattice with N particles.

$$\sum_i \Phi_1 \begin{pmatrix} i \\ \alpha \end{pmatrix} = 0 \qquad \ldots (5.1)$$

$$\sum_i \Phi_1 \begin{pmatrix} i \\ \alpha \end{pmatrix} R \begin{pmatrix} i \\ \beta \end{pmatrix} \quad \text{symmetric in } \alpha, \beta \qquad \ldots (5.2)$$

$$\sum_j \Phi_2 \begin{pmatrix} ij \\ \alpha\beta \end{pmatrix} = 0 \qquad \ldots (5.3)$$

$$\sum_j \Phi_2 \begin{pmatrix} ij \\ \alpha\gamma \end{pmatrix} R \begin{pmatrix} j \\ \beta \end{pmatrix} + \Phi_1 \begin{pmatrix} i \\ \gamma \end{pmatrix} \delta_{\alpha\beta} \quad \text{symmetric in } \beta, \gamma \qquad \ldots (5.4)$$

where $R \begin{pmatrix} i \\ \alpha \end{pmatrix} = \alpha$-th component of $\boldsymbol{R}_i$ etc.

The first two equations give the expected six relations between the $3N$ first order e.p.'s. The third equation apparently gives $9N$ relations corresponding to $9N$ values of (i, α, β). But all of them are not independent From the definition of Φ_2 follows the identity

$$\sum_{ij} \Phi_2 \begin{pmatrix} ij \\ \alpha\beta \end{pmatrix} = \sum_{ij} \Phi_2 \begin{pmatrix} ij \\ \beta\alpha \end{pmatrix} \quad \dots \quad (6)$$

which is not redundant for $\alpha \neq \beta$. Hence three (for $\alpha \neq \beta$) of the relations given by eq (5.3) are not independent Thus the total number of independent relations obtained from eq. (5.3) is $9N-3$.

The relation (5.4) is not redundant for $\beta \neq \gamma$ We consider two cases. For the case $\alpha = \gamma \neq \beta$ (or $\alpha = \beta \neq \gamma$)(5.4) is of the form

$$\sum_j \Phi_2 \begin{pmatrix} ij \\ \alpha\alpha \end{pmatrix} R \begin{pmatrix} j \\ \beta \end{pmatrix} = \sum_j \Phi_2 \begin{pmatrix} ij \\ \alpha\beta \end{pmatrix} R \begin{pmatrix} j \\ \alpha \end{pmatrix} + \Phi_1 \begin{pmatrix} i \\ \beta \end{pmatrix} \quad \dots \quad (7)$$

There are apparently $6N$ relations for $6N$ possible values of $(i, \alpha, \beta)(\alpha \neq \beta)$. But on summing both sides of eq (7) with respect to i each side becomes zero because of eqs. (5.1) and (5.3) Hence for each α, β one relation of the type of eq (7) becomes dependent and we have $6N-6$ independent relations of this type.

The second case arises when in (5.4) $\alpha \neq \beta \neq \gamma$. There are apparently $3N$ relations of the form

$$\sum_j \Phi_2 \begin{pmatrix} ij \\ \alpha\gamma \end{pmatrix} R \begin{pmatrix} j \\ \beta \end{pmatrix} = \sum_j \Phi_2 \begin{pmatrix} ij \\ \alpha\beta \end{pmatrix} R \begin{pmatrix} j \\ \gamma \end{pmatrix} \quad \dots \quad (8)$$

Here also on summing over i each side of (8) becomes zero. So three of the relations drop out giving apparently $3N-3$ relations of the type of eq (8). Again using eqs. (7) and (5.2) we get for $\alpha \neq \beta \neq \gamma$

$$\sum_{ij} \Phi_2 \begin{pmatrix} ij \\ \alpha\beta \end{pmatrix} R \begin{pmatrix} j \\ \alpha \end{pmatrix} R \begin{pmatrix} i \\ \gamma \end{pmatrix} = \sum_{ij} \Phi_2 \begin{pmatrix} ij \\ \alpha\gamma \end{pmatrix} R \begin{pmatrix} j \\ \alpha \end{pmatrix} R \begin{pmatrix} i \\ \beta \end{pmatrix} \quad \dots \quad (9)$$

Starting from the identity

$$\sum_{ij} \Phi_2 \begin{pmatrix} ij \\ \gamma\beta \end{pmatrix} R \begin{pmatrix} i \\ \alpha \end{pmatrix} R \begin{pmatrix} j \\ \alpha \end{pmatrix} = \sum_{ij} \Phi_2 \begin{pmatrix} ij \\ \beta\gamma \end{pmatrix} R \begin{pmatrix} i \\ \alpha \end{pmatrix} R \begin{pmatrix} j \\ \alpha \end{pmatrix} \quad \dots \quad (9.1)$$

We can independently obtain the relation (9) by using the relation (8) Thus all the $3N-3$ relations in eq. (8) are not independent. Since there are three relations of the form of eq. (9) the number of independent relations in (8) will be $3N$-6. ultimately we get $(6N-6)+(3N-6) = 9N-12$ independent relations from relation (5.4).

The total number of independent invariance conditions obtained from eqs. (5.3) and (5.4) is, therefore, $(9N-3)+(9N-12) = 18N-15$. As already pointed out this is the number of relations necessary to guarantee the translational and rotational invariance of energy expression (2)

Thus we find that the B-H invariance conditions (5.1) to (5.4) are complete and sufficient

3. Remarks

It should be noted that equilibrium conditions such as the vanishing of tension in a linear chain or vanishing of stresses in a lattice will put some additional restrictions on the first order c.p.'s which through rotational invariance conditions (5.1) will put restrictions on the second order c.p.'s

It can be shown (Sarkar and Sengupta) that the additional conditions obtained by some workers mentioned in the introduction can be obtained by imposing equilibrium conditions rather than using the unusual procedure of going to second order terms in rotation parameter when considering the invariance of potential energy

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